

FORM PTO-1390  
(REV. 9-2001)

U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE

ATTORNEY'S DOCKET NUMBER

TRANSMITTAL LETTER TO THE UNITED STATES  
DESIGNATED/ELECTED OFFICE (DO/EO/US)  
CONCERNING A FILING UNDER 35 U.S.C. 371

PST 6221

U.S. APPLICATION NO. (If known, see 37 CFR 1.5

10/018871

INTERNATIONAL APPLICATION NO.	INTERNATIONAL FILING DATE	PRIORITY DATE CLAIMED
PCT/SE00/01285	19 June 2000	29 June 1999

TITLE OF INVENTION  
**COMPOUNDS FROM EPOXIDISED NITRILES, PROCESS FOR THEIR PRODUCTION AND USE AS  
CLEANING AGENTS**APPLICANT(S) FOR DO/EO/US  
**Elina Sandberg and Eva Gottberg-Klingskog**

Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1.  This is a **FIRST** submission of items concerning a filing under 35 U.S.C. 371.
2.  This is a **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. 371.
3.  This is an express request to begin national examination procedures (35 U.S.C. 371(f)). The submission must include items (5), (6), (9) and (21) indicated below.
4.  The US has been elected by the expiration of 19 months from the priority date (Article 31).
5.  A copy of the International Application as filed (35 U.S.C. 371(c)(2))
  - a.  is attached hereto (required only if not communicated by the International Bureau).
  - b.  has been communicated by the International Bureau.
  - c.  is not required, as the application was filed in the United States Receiving Office (RO/US).
6.  An English language translation of the International Application as filed (35 U.S.C. 371(c)(2))
  - a.  is attached hereto.
  - b.  has been previously submitted under 35 U.S.C. 154(d)(4).
7.  Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3))
  - a.  are attached hereto (required only if not communicated by the International Bureau).
  - b.  have been communicated by the International Bureau.
  - c.  have not been made; however, the time limit for making such amendments has NOT expired.
  - d.  have not been made and will not be made.
8.  An English language translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).
9.  An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)). (**unsigned**)
10.  An English language translation of the annexes of the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).

**Items 11 to 20 below concern document(s) or information included:**

11.  An Information Disclosure Statement under 37 CFR 1.97 and 1.98.
12.  An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.
13.  A **FIRST** preliminary amendment. **Marked Up Version of Specification and Claims**
14.  A **SECOND** or **SUBSEQUENT** preliminary amendment.
15.  A substitute specification.
16.  A change of power of attorney and/or address letter.
17.  A computer-readable form of the sequence listing in accordance with PCT Rule 13ter.2 and 35 U.S.C. 1.821 - 1.825.
18.  A second copy of the published international application under 35 U.S.C. 154(d)(4).
19.  A second copy of the English language translation of the international application under 35 U.S.C. 154(d)(4).
20.  Other items or information: **Copy of International Search Report dated October 11, 2000**  
**Copy of International Preliminary Examination Report**  
**dated June 12, 2000**  
**Express Mail Label No: EM122093188US**

U.S. APPLICATION NO. (if known, see 37 CFR 1.5) <b>10/018879</b>	INTERNATIONAL APPLICATION NO. <b>PCT/SE00/01285</b>	ATTORNEY'S DOCKET NUMBER <b>PST 6221</b>																				
<p><input type="checkbox"/> The following fees are submitted:</p> <p><b>BASIC NATIONAL FEE (37 CFR 1.492 (a) (1) - (5)):</b></p> <p>Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO and International Search Report not prepared by the EPO or JPO ..... <b>\$1040.00</b></p> <p>International preliminary examination fee (37 CFR 1.482) not paid to USPTO but International Search Report prepared by the EPO or JPO ..... <b>\$890.00</b></p> <p>International preliminary examination fee (37 CFR 1.482) not paid to USPTO but international search fee (37 CFR 1.445(a)(2)) paid to USPTO ..... <b>\$740.00</b></p> <p>International preliminary examination fee (37 CFR 1.482) paid to USPTO but all claims did not satisfy provisions of PCT Article 33(1)-(4) ..... <b>\$710.00</b></p> <p>International preliminary examination fee (37 CFR 1.482) paid to USPTO and all claims satisfied provisions of PCT Article 33(1)-(4) ..... <b>\$100.00</b></p>		<b>CALCULATIONS PTO USE ONLY</b>																				
<b>ENTER APPROPRIATE BASIC FEE AMOUNT =</b>		<b>\$1040.00</b>																				
<p>Surcharge of <b>\$130.00</b> for furnishing the oath or declaration later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(e)).</p> <table border="1" style="width: 100%; border-collapse: collapse;"> <thead> <tr> <th>CLAIMS</th> <th>NUMBER FILED</th> <th>NUMBER EXTRA</th> <th>RATE</th> <th>\$</th> </tr> </thead> <tbody> <tr> <td>Total claims</td> <td><b>15</b> - 20 =</td> <td><b>0</b></td> <td>x <b>\$18.00</b></td> <td>\$ <b>—</b></td> </tr> <tr> <td>Independent claims</td> <td><b>2</b> - 3 =</td> <td><b>0</b></td> <td>x <b>\$84.00</b></td> <td>\$ <b>—</b></td> </tr> <tr> <td>MULTIPLE DEPENDENT CLAIM(S) (if applicable)</td> <td></td> <td></td> <td>+ <b>\$280.00</b></td> <td>\$ <b>—</b></td> </tr> </tbody> </table>		CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE	\$	Total claims	<b>15</b> - 20 =	<b>0</b>	x <b>\$18.00</b>	\$ <b>—</b>	Independent claims	<b>2</b> - 3 =	<b>0</b>	x <b>\$84.00</b>	\$ <b>—</b>	MULTIPLE DEPENDENT CLAIM(S) (if applicable)			+ <b>\$280.00</b>	\$ <b>—</b>	
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<b>TOTAL OF ABOVE CALCULATIONS =</b>		<b>\$1040.00</b>																				
<p><input type="checkbox"/> Applicant claims small entity status. See 37 CFR 1.27. The fees indicated above are reduced by 1/2.</p>		+																				
		<b>SUBTOTAL =</b>	<b>\$1040.00</b>																			
<p>Processing fee of <b>\$130.00</b> for furnishing the English translation later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(f)).</p>																						
		<b>TOTAL NATIONAL FEE =</b>	<b>\$1040.00</b>																			
<p>Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). <b>\$40.00</b> per property</p>		+	\$																			
		<b>TOTAL FEES ENCLOSED =</b>	<b>\$</b>																			
		<b>Amount to be refunded:</b>	\$																			
		<b>charged:</b>	<b>\$1040.00</b>																			
<p>a. <input type="checkbox"/> A check in the amount of \$ _____ to cover the above fees is enclosed.</p> <p>b. <input checked="" type="checkbox"/> Please charge my Deposit Account No. <b>01-1350</b> in the amount of <b>\$1040.00</b> to cover the above fees. A duplicate copy of this sheet is enclosed.</p> <p>c. <input checked="" type="checkbox"/> The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. <b>01-1350</b>. A duplicate copy of this sheet is enclosed.</p> <p>d. <input type="checkbox"/> Fees are to be charged to a credit card. <b>WARNING:</b> Information on this form may become public. <b>Credit card information should not be included on this form.</b> Provide credit card information and authorization on PTO-2038.</p>																						
<p><b>NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137 (a) or (b)) must be filed and granted to restore the application to pending status.</b></p>																						
<p>SEND ALL CORRESPONDENCE TO:</p> <p style="text-align: right;"> SIGNATURE</p> <p style="text-align: right;"><b>Ralph J. Mancini</b> NAME</p> <p style="text-align: right;"><b>34,054</b> REGISTRATION NUMBER</p>																						

**PATENT****IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**

Application of:

SANDBERG, Elina

Docket No.: PST6221

Serial No.: Unassigned

Int'l Application No.: PCT/SE00/01285

Group Art Unit:

Int'l Filing Date: June 19, 2000

Priority Date: June 29, 1999

Examiner:

Title: COMPOUNDS FROM EPOXIDISED NITRILES:  
PROCESS FOR THEIR PRODUCTION AND  
USE AS CLEANING AGENTSAssistant Commissioner of Patents  
Washington, D.C. 20231**PRELIMINARY AMENDMENT**

Sir:

In accordance with the provisions of 37 C.F.R. §1.111, applicants provide the following amendments and remarks for entry in the above-identified case.

**IN THE SPECIFICATION**Please amend the specification as follows:

At page 1, after the Title, please insert the following:

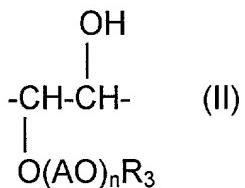
--The present application was filed on June 19, 2000 as international application serial number PCT/SE00/01285 and claims priority of Swedish patent application No. 99002450-7 filed on June 29, 1999.--

**IN THE CLAIMS**

Please cancel claims 2-10.

Please amend the claims as follows:

1. Nonionic compounds of the general formula RY (I), where R is a substituted aliphatic group containing 1-3 structure elements of the formula

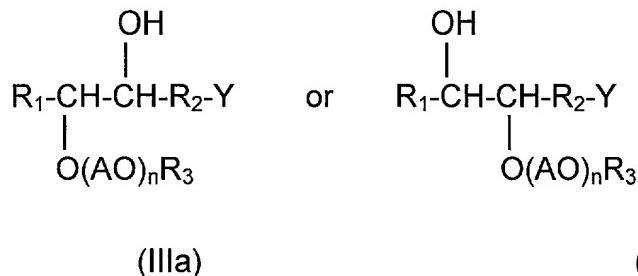


where the carbon atoms shown in the structure element are part of the aliphatic carbon skeleton of group R, which contains 8-24 carbon atoms, and Y is a nitrile or an amide group; R<sub>3</sub> is an alkyl group with 1-4 carbon atoms; AO is an alkyleneoxy group containing 2-4 carbon atoms and n is a number between 1 and 30.

Please add new claims 11-25.

- 11. Nonionic compounds of claim 1 containing 1-2 structure elements according to formula (II). --

- 12. Nonionic compounds of the general formulae

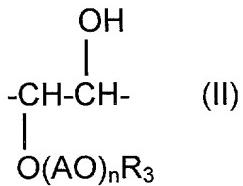


where R<sub>1</sub> is an aliphatic group, R<sub>2</sub> is an aliphatic radical, the sum of carbon atoms contained in R<sub>1</sub> and R<sub>2</sub> is between 9 and 19; Y is a nitrile or an amide group; R<sub>3</sub> is an alkyl group with 1-4 carbon atoms; AO is an alkyleneoxy group containing 2-4 carbon atoms and n is a number between 1 and 30.--

- 13. Nonionic compounds according to claim 1 where at least 50% of the AO groups are ethyleneoxy groups.--
- 14. Nonionic compounds according to claim 2 where at least 50% of the AO groups are ethyleneoxy groups.--
- 15. Nonionic compounds according to claim 3 where at least 50% of the AO groups are ethyleneoxy groups.--
- 16. Nonionic compounds according to claim 1 where the AO group is the ethyleneoxy group.--
- 17. Nonionic compounds according to claim 2 where the AO group is the ethyleneoxy group.--
- 18. Nonionic compounds according to claim 3 where the AO group is the ethyleneoxy group.--
- 19. Nonionic compounds according to claim 1 where n is 3-20 and R<sub>3</sub> is methyl or ethyl.--
- 20. Nonionic compounds according to claim 2 where the AO group is the ethyleneoxy group.--
- 21. Nonionic compounds according to claim 3 where the AO group is the ethyleneoxy group.--

--22. A method of producing the nonionic compound of claim 1 which comprises  
a) reacting an epoxidised nitrile containing 1-3 epoxy groups and a total of 8 to 24 carbon atoms with an alkyl blocked polyalkylene glycol having the formula  $R_3O(AO)_nH$ , where  $R_3$  is an alkyl group with 1-4 carbon atoms; AO is an alkyleneoxy group containing 2-4 carbon atoms and n is a number between 1 and 30, in the presence of a catalyst, and optionally subjecting the product obtained to alkaline hydrogen peroxide.

--23. A method of producing the nonionic compound of claim 1 which comprises reacting ammonia or a primary or secondary amine with an acid or an ester containing 1-3 structure elements according to formula II



where  $R_3$  is an alkyl group with 1-4 carbon atoms; AO is an alkyleneoxy group containing 2-4 carbon atoms and n is a number between 1 and 30.--

--24. A surfactant composition which comprises an effective amount of at least one non-ionic compound of claim 1. --

--25. The surfactant composition of claim 23 adapted for the cleaning of hard surfaces, vehicle cleaning, bottle cleaning, machine dishwashing or machine washing of textiles.--

#### Remarks

This is an international application filed under the Patent Cooperation Treaty (PCT) on June 19, 2000. The examiner is respectfully requested to note that prior to the present amendment, claims 1-10 were pending in the present application. In the present amendment, claims 2-10 are cancelled and new claims 11-25 are added to the application. New claims 11-25 correspond substantially with the subject matter of

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cancelled claims 2-10. No new issues are raised by the amendments and it is believed that the amendments have placed the pending claims are in ideal condition for U.S. prosecution.

The Examiner is reminded that the present application was filed under the PCT and that Unity of Invention rules apply.

Attached hereto is a marked-up version of the changes made to the specification and claims by the current amendment. The attached page is captioned "Version with markings to show changes made."

Since the present amendment raises no new issues and presents no new matter, entry thereof in accordance with 37 C.F.R. §1.111 prior to the initial examination of the present case on the merits is respectfully requested.

Respectfully submitted,



Ralph J. Mancini  
Attorney for Applicants  
Registration No. 34,054

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Intellectual Property Department  
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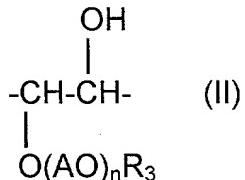
**Version with markings to show changes made**

The following sentence was added on page 1 of the specification after the title:

--The present application was filed on June 19, 2000 as international application serial number PCT/SE00/01285 and claims priority of Swedish patent application No. 99002450-7 filed on June 29, 1999.--

Claim 1 was amended as follows:

1. Nonionic compounds of characterised by the general formula RY (I), where R is a substituted aliphatic group containing 1-3 structure elements with of the formula

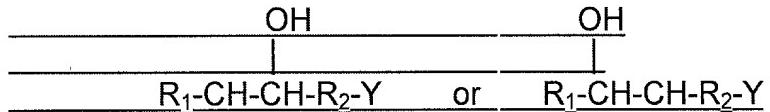


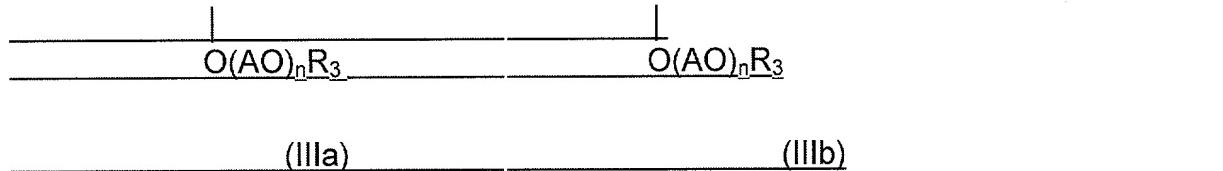
where the carbon atoms shown in the structure element are part of the aliphatic carbon skeleton of group R, which contains 8-24 carbon atoms, and Y is a nitrile or an amide group; R<sub>3</sub> is an alkyl group with 1-4 carbon atoms; AO is an alkyleneoxy group containing 2-4 carbon atoms and n is a number between 1 and 30.

The following new claims are added to the application:

--11. Nonionic compounds of claim 1 containing 1-2 structure elements according to formula (II). --

--12. Nonionic compounds of the general formulae





where  $\text{R}_1$  is an aliphatic group,  $\text{R}_2$  is an aliphatic radical, the sum of carbon atoms contained in  $\text{R}_1$  and  $\text{R}_2$  is between 9 and 19; Y is a nitrile or an amide group;  $\text{R}_3$  is an alkyl group with 1-4 carbon atoms; AO is an alkyleneoxy group containing 2-4 carbon atoms and n is a number between 1 and 30.--

--13. Nonionic compounds according to claim 1 where at least 50% of the AO groups are ethyleneoxy groups.--

--14. Nonionic compounds according to claim 2 where at least 50% of the AO groups are ethyleneoxy groups.--

--15. Nonionic compounds according to claim 3 where at least 50% of the AO groups are ethyleneoxy groups.--

--16. Nonionic compounds according to claim 1 where the AO group is the ethyleneoxy group.--

--17. Nonionic compounds according to claim 2 where the AO group is the ethyleneoxy group.--

--18. Nonionic compounds according to claim 3 where the AO group is the ethyleneoxy group.--

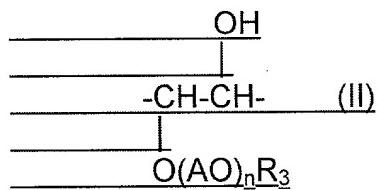
--19. Nonionic compounds according to claim 1 where n is 3-20 and  $\text{R}_3$  is methyl or ethyl.--

--20. Nonionic compounds according to claim 2 where the AO group is the ethyleneoxy group.--

--21. Nonionic compounds according to claim 3 where the AO group is the ethyleneoxy group.--

--22. A method of producing the nonionic compound of claim 1 which comprises  
a) reacting an epoxidised nitrile containing 1-3 epoxy groups and a total of 8 to 24 carbon atoms with an alkyl blocked polyalkylene glycol having the formula  $R_3O(AO)_nH$ , where  $R_3$  is an alkyl group with 1-4 carbon atoms; AO is an alkyleneoxy group containing 2-4 carbon atoms and n is a number between 1 and 30, in the presence of a catalyst, and optionally subjecting the product obtained to alkaline hydrogen peroxide.

--23. A method of producing the nonionic compound of claim 1 which comprises reacting ammonia or a primary or secondary amine with an acid or an ester containing 1-3 structure elements according to formula II



where  $R_3$  is an alkyl group with 1-4 carbon atoms; AO is an alkyleneoxy group containing 2-4 carbon atoms and n is a number between 1 and 30.--

--24. A surfactant composition which comprises an effective amount of at least one non-ionic compound of claim 1. --

--25. The surfactant composition of claim 23 adapted for the cleaning of hard surfaces, vehicle cleaning, bottle cleaning, machine dishwashing or machine washing of textiles.--

COMPOUNDS FROM EPOXIDISED NITRILES, PROCESS FOR THEIR  
PRODUCTION AND USE AS CLEANING AGENTS

The present invention relates to nonionic compounds  
of the polyoxyalkylene type that are low-foaming and can be  
used as surfactants, especially in cleaning compositions at  
an alkaline pH. They are obtainable from unsaturated  
nitriles, that have been epoxidised with e.g. hydrogen  
peroxide, and alkyl blocked polyalkylene glycols. The nitrile  
surfactants can be reacted further with e.g. hydrogen  
peroxide under alkaline conditions to obtain amides. Amide  
derivatives of this type can also be obtained from  
unsaturated acids or esters that have been epoxidised and  
thereafter reacted with alkyl blocked polyalkylene glycols.  
These acid or ester derivatives are transformed to the  
corresponding amide derivatives by reaction with ammonia or a  
primary or secondary amine.

Nonionic surfactants constitute an important group of  
surface-active compounds that are widely used in numerous  
applications. However, many of the ethylene oxide adducts  
containing a hydrophobic chain with 12-22 carbon atoms are  
too high-foaming to be used in certain applications, such as  
e.g. machine dish-washing, machine washing of textiles,  
bottle cleaning and cleaning of hard surfaces.

It has been suggested in EP-A2-0 754 667 to produce  
new nonionic surfactants, that would generate less foam, by  
reacting an epoxidised fatty acid ester with a polyglycol  
ether. These ring-opened products are claimed to be  
particularly suited to be used as defoamers in a variety of  
applications. In WO 94/07840 similar products are obtained by  
reacting epoxidised esters with fatty alcohol polyglycol  
ethers. These products are claimed to be low-foaming and to  
have a good biodegradability, and are used as auxiliaries in  
the removal of water from solid materials.

However, there is a drawback when using the ester  
polyglycol ether compounds, since the ester group is readily  
hydrolysed when subjected to highly alkaline conditions. This

excludes applications where a high pH is required, since the nonionic product would then be converted to an anionic product.

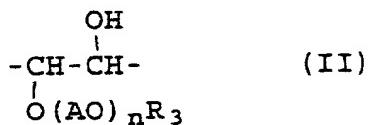
Nonionic products containing a cyano group, which is a more stable functional group than the ester group, have been described by Wrigley, Smith and Stirton (J. Am. Oil Chem. Soc. 39:80-84 (1962)). These surfactants were obtained by treatment of an unsaturated nitrile with 98% formic acid and 30% hydrogen peroxide to give the hydroxy-formate, followed by mild hydrolysis to yield the dihydroxy compound, which was then ethoxylated using KOH as a catalyst. However, since both the hydroxyl groups of the intermediate product are secondary, the final product will not be very well defined. Once an ethylene oxide unit has been attached to the secondary hydroxyl group, there will be a primary hydroxyl group present, which will add another ethylene oxide unit much faster than the remaining secondary groups. The end result will be a product where one of the polyoxyethylene chains is much longer than the other, a lot of the polyoxyethylene homologues will probably contain only short chains, and some part of the hydrophobic diol will even be unreacted. Thus, in the above-mentioned article it is stated that ethoxylation of 9,10-dihydroxystearonitrile with 4 moles of ethylene oxide leaves 5.9 % of starting diol.

Consequently there is a need for low-foaming products that are more stable than the prior known ester polyglycol ethers and better defined than the ethoxylated diol nitriles. The purpose of the present invention is to provide such products.

It has now been found that these objectives can be met by a new class of low-foaming well-defined compounds, which can easily be prepared from starting materials that are easy to handle and that can be obtained by standard procedures, by reacting an epoxidised nitrile with an alkyl blocked polyalkylene glycol. Since the compounds that are obtained are low-foaming, they are suitable to use e.g. in cleaning compositions for applications where low foam is

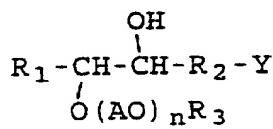
required, such as vehicle cleaning, bottle cleaning and machine-washing. To obtain products with even better toxicological and environmental profiles that still are relatively stable, the nitrile group can be converted to an amide group by treatment with e.g. alkaline hydrogen peroxide. An amide derivative according to the invention could also be produced by the reaction between an acid or ester derivative of the type described in EP-A2-0 754 667 and ammonia or a primary or secondary amine.

The nonionic compounds according to the present invention are characterised by the general formula RY (I), where R is a substituted aliphatic group containing 1-3 structure elements with the formula

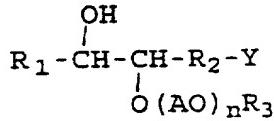


where the carbon atoms shown in the structure element are part of the aliphatic carbon skeleton of group R, which contains 8-24 carbon atoms, preferably 12-22 carbon atoms, and Y is a nitrile or an amide group; R<sub>3</sub> is an alkyl group with 1-4 carbon atoms; AO is an alkyleneoxy group containing 2-4 carbon atoms and n is a number between 1 and 30, preferably 3-20. The number of structure elements contained in the chain is preferably 1-2.

Suitable examples of the above-mentioned compounds are those having the formulae



or

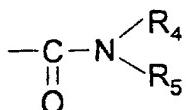


(IIIa)

(IIIb)

where R<sub>1</sub> is an aliphatic group, R<sub>2</sub> is an aliphatic radical, the sum of carbon atoms contained in R<sub>1</sub> and R<sub>2</sub> is between 9 and 19 and R<sub>3</sub>, AO, n and Y have the same meaning as above.

Suitable amide groups are those having the formula



where  $\text{R}_4$  and  $\text{R}_5$  independently are H, an alkyl group with 1-6 carbon atoms, preferably 1-4 carbon atoms, or  $(\text{AO})_m$  where AO is an alkyleneoxy group containing 2-3 carbon atoms, preferably 2, and m is 1-20, preferably 1-2. Most preferably at least one of  $\text{R}_4$  and  $\text{R}_5$  is H.

The nonionic compounds according to the invention may be produced by

- a) reacting an epoxidised nitrile containing 1-3 epoxy groups and a total of 8 to 24 carbon atoms, preferably 12-22 carbon atoms, with an alkyl blocked polyalkylene glycol having the formula  $\text{R}_3\text{O}(\text{AO})_n\text{H}$ , where  $\text{R}_3$ , AO and n has the same meaning as in formula II, in the presence of a catalyst, and optionally subjecting the product obtained to alkaline hydrogen peroxide or
- b) reacting ammonia or a primary or secondary amine with an acid or an ester containing 1-3 structure elements according to formula II in the aliphatic skeleton. The alkyleneoxy groups could be distributed randomly or in blocks or a mixture thereof. Normally at least 50% of the alkyleneoxy groups are ethyleneoxy groups. Alkyl blocked polyethylene glycols are the reactants most often used. Suitable examples of alkyl blocked polyalkylene glycols are  $\text{CH}_3(\text{CH}_2\text{CH}(\text{CH}_3)\text{O})_2(\text{CH}_2\text{CH}_2\text{O})_{10}\text{H}$  and  $\text{CH}_3(\text{CH}_2\text{CH}_2\text{O})_{14}\text{H}$ .

When the reaction between a monoepoxidised nitrile and the alkyl blocked polyalkylene glycol takes place, there is an equal chance for attack by the glycol on either of the two carbons in the epoxy ring. Consequently two regioisomers are formed in about the same amount, as is shown by formula IIIa and IIIb.

The epoxidised nitriles could be derived from unsaturated nitriles e.g. by using the Venturello method described in GB-A-2055821, where hydrogen peroxide is used as the source of active oxygen, or by the use of peracids. The unsaturated nitriles could contain more than one double bond,

and the epoxidation of such polyunsaturated nitriles can consequently give rise to products containing more than one epoxy group. Accordingly, the nonionic compounds derived from such polyepoxidised nitriles could contain several  
5 polyoxyethylene chains distributed along the aliphatic skeleton. Naturally occurring fatty acids, which are the starting material for the nitriles, are normally mixtures between acids with different amounts of double bonds, and consequently the resulting end products will also be mixtures  
10 of compounds containing different numbers of polyoxyethylene chains.

The unsaturated nitriles are intermediates in the process for the manufacturing of fatty amines, and they are thus readily available in large quantities. Oleonitrile is a suitable example of an unsaturated nitrile, but also other nitriles derived from unsaturated acids would be possible to use. Examples of such acids are tall oil acid, linoleic acid, erucic acid and fish-oil acids, as well as unsaturated  
15 synthetic acids.

The reaction between the epoxynitriles and the alkyl blocked polyalkylene glycols could be catalysed both by acids, such as Lewis acids e.g.  $\text{BF}_3$ , and by alkali, such as  
20 alkoxides.

The conversion of the nitrile group to an amide group by the treatment with alkaline peroxide is described e.g. in  
25 Organic Synthesis Collective Vol. II p. 586.

The nonionic amide compounds according to the invention could also be obtained by a standard procedure via the suitable acid or ester derivatives by reaction with  
30 ammonia or an amine. Suitable examples of amines are primary or secondary alkylamines, ethanolamine and diethanolamine.

The following examples are illustrative of the invention and are not to be construed as limiting thereof.

#### Example A

35 The product mixture according to formula IIIa and IIIb where  $R_1=\text{C}_8\text{H}_{17}$ ,  $R_2=\text{C}_7\text{H}_{14}$ ,  $\text{AO}=-\text{CH}_2\text{CH}_2\text{O}-$ ,  $n=10$ ,  $R_3=\text{CH}_3$  and  $Y=-\text{CN}$  was prepared according to the following procedure:

0.01 mole boron trifluoride diethyl ether complex was added to 0.1 mole polyethylene glycol monomethyl ether (molecular weight = 472.5) while stirring. The mixture was heated to 60°C, and with continued stirring 0.1 mole 9,10-epoxyoleonitrile was added during a period of 30 minutes. The reaction was exothermic, and the temperature was kept between 60-65°C during the addition by alternately cooling and heating the reaction mixture. When all epoxide had been added, a sample was removed to determine the amount of unreacted epoxide present in the product. The method used for this analysis was a titration procedure, which is described in Analytical Chemistry 36 (1964) p667. The amount of epoxide was <1%. The product was also investigated by  $^1\text{H-NMR}$ , and the spectrum obtained was in accordance with the desired structure.

Sodium bicarbonate was added to the product in order to remove  $\text{BF}_3$ . The slurry obtained was stirred for 15 minutes, and then filtered through a layer of sodium bicarbonate.

**Example B**

The product mixture according to formula IIIa and IIIb where  $R_1=\text{C}_8\text{H}_{17}$ ,  $R_2=\text{C}_7\text{H}_{14}$ ,  $\text{AO}=-\text{CH}_2\text{CH}_2\text{O}-$ ,  $n=14$ ,  $R_3=\text{CH}_3$  and  $\text{Y}=-\text{CN}$  was prepared according to the procedure described in Example A, except that the glycol used was polyethylene glycol monomethyl ether with a molecular weight of 649. The amount of unreacted epoxide in the product mixture was <1%, and the  $^1\text{H-NMR}$  spectrum was in accordance with the desired structure.

**Example C**

The product mixture according to formula IIIa and IIIb where  $R_1=\text{C}_8\text{H}_{17}$ ,  $R_2=\text{C}_7\text{H}_{14}$ ,  $\text{AO}=-\text{CH}_2\text{CH}_2\text{O}-$ ,  $n=7.2$ ,  $R_3=\text{CH}_3$  and  $\text{Y}=-\text{CN}$  was prepared according to the procedure described in Example A, except that the glycol used was polyethylene glycol monomethyl ether with a molecular weight of 350. The amount of unreacted epoxide in the product mixture was <1%, and the  $^1\text{H-NMR}$  spectrum was in accordance with the desired structure.

**Example D**

The product mixture according to formula IIIa and IIIb where  $R_1=C_8H_{17}$ ,  $R_2=C_7H_{14}$ ,  $AO=-CH_2CH_2O-$ ,  $n=11.8$ ,  $R_3=CH_3$  and  $Y=-CN$  was prepared according to the procedure described in Example A, except that the glycol used was polyethylene glycol monomethyl ether with a molecular weight of 550. The amount of unreacted epoxide in the product mixture was <1%, and the  $^1H$ -NMR spectrum was in accordance with the desired structure.

**Example E**

To 35.7 g (0.052 moles) of the product mixture from example C was added 135 ml of acetone and 1 ml of Dequest 2006 (complexing agent). The mixture was heated to 60°C and during a period of 30 minutes 8.7 g of 6M KOH solution in water and 9.1 g (0.080 moles) of 30%  $H_2O_2$  was added dropwise to the solution. After the addition the reaction mixture was kept at 60°C for 3 hours. An analysis by  $^1H$ -NMR revealed that 29% of the nitrile groups had been converted to primary amide groups.

**Example 1**

The foaming behaviour of an epoxidised oleonitrile that had been reacted with a methyl-blocked polyethylene glycol containing 10 (example A) and 14 (example B) polyoxyethylene units respectively, was studied. The foam was measured as mm foam produced in a 500 ml measuring cylinder with 49 mm inner diameter from 200 ml 0.5% surfactant solution when the cylinder was turned around 40 times in one minute. The test was made at room temperature, and the foam height was registered directly and after 1, 5 and 10 minutes. Ethoxylated dihydroxystearonitrile, nonylphenol that had been ethoxylated with 10 moles of ethylene oxide (NF + 10 EO) and  $C_{12}-C_{16}$  alcohol that had been ethoxylated with 7.5 moles of ethylene oxide were used as references. The latter two products are well-known surfactants that are used in cleaning processes.

Product	Foam height (mm) after 0 min	Foam height (mm) after 1 min	Foam height (mm) after 5 min	Foam height (mm) after 10 min
Example A	86	51	3	1-2
Example B	110	85	11	7
Dihydroxy-stearo-nitrile + 10 EO	155	120	7	5
Dihydroxy-stearo-nitrile + 14 EO	172	141	13	10
Example C	58	19	8	5
Example D	104	100	23	11
NF + 10EO	>230	>230	>230	>230
C <sub>12</sub> -C <sub>16</sub> alcohol + 7.5 EO	220	217	203	176

The products according to the present invention produce less foam than the corresponding 9,10-dihydroxystearonitrile derivatives containing the same amount of oxyethylene units. NF + 10EO and C<sub>12</sub>-C<sub>16</sub> alcohol + 7.5 EO produce much more foam than any of the products investigated.

**Example 2**

The following recipe was used to prepare formulations containing the surfactants according to the present invention.

Component	% by weight of component
Nonionic surfactant	5
Tetrapotassium pyrophosphate	6
Sodium metasilicate x 5 H <sub>2</sub> O	4
Hydrotrope (hexyl glucoside)	variable <sup>1</sup>
Water	balance

<sup>1</sup>the amount of hydrotrope added is the minimum amount needed to obtain a clear solution at room temperature

The cleaning efficiency of the formulations was evaluated using the following cleaning test: White painted plates were smeared with an oil-soot mixture obtained from diesel engines. 25 ml of the test solutions were poured onto the top of the oil-smeared plates and left there for one minute. The plates were then rinsed off with a rich flow of water. All solutions and the water were kept at a temperature of about 15-20°C. All test solutions were placed on the same plate. The reflectance of the plates was measured with a Minolta Chroma Meter CR-200 reflectometer before and after cleaning.

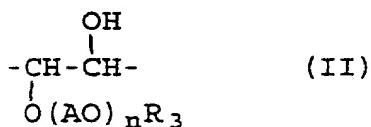
The test was performed with solutions diluted 1:10 with water. The washed-away soil was calculated by the computer program integrated in the meter, and the results are collected in the table below. One value was obtained for each substance by calculating the mean of four measurements obtained at four different spots on the plate. The values in the table are the mean of the values obtained on two different plates. The reference is a traditional nonionic surfactant obtained by ethoxylation of a C12-C16 alcohol with 7.5 moles of ethylene oxide.

Surfactant in formulation	% washed away soil with 1:10 dilution
Example C	75
Example D	86
C12-C16 alcohol + 7.5 EO	76

As can be seen from the table, the compounds of the present invention are as good cleaners as the traditional nonionic surfactant used as a reference, and they also have the advantage of being more low-foaming.

## C L A I M S

1. Nonionic compounds characterised by the general formula RY (I), where R is a substituted aliphatic group containing 1-3 structure elements with the formula



where the carbon atoms shown in the structure element are part of the aliphatic carbon skeleton of group R, which contains 8-24 carbon atoms, and Y is a nitrile or an amide group; R<sub>3</sub> is an alkyl group with 1-4 carbon atoms; AO is an alkyleneoxy group containing 2-4 carbon atoms and n is a number between 1 and 30.

2. Nonionic compounds according to claim 1 containing 1-2 structure elements according to formula (II).

3. Nonionic compounds characterised by the general formulae



(IIIa) (IIIb)

where R<sub>1</sub> is an aliphatic group, R<sub>2</sub> is an aliphatic radical, the sum of carbon atoms contained in R<sub>1</sub> and R<sub>2</sub> is between 9 and 19; and R<sub>3</sub>, AO, n and Y have the same meaning as in claim 1.

4. Nonionic compounds according to claims 1-3 where at least 50% of the AO groups are ethyleneoxy groups.

5. Nonionic compounds according to claims 1-3 where the AO group is the ethyleneoxy group.

6. Nonionic compounds according to claims 1-3 where n is 3-20 and R<sub>3</sub> is methyl or ethyl.

7. A method of producing a nonionic compound according to claims 1-6, characterised in that the compound is produced by

- 5           a)       reacting an epoxidised nitrile containing 1-3 epoxy groups and a total of 8 to 24 carbon atoms with an alkyl blocked polyalkylene glycol having the formula  $R_3O(AO)_nH$ , where  $R_3$ , AO and n has the same meaning as in claims 1-6, in the presence of a catalyst, and optionally subjecting the product obtained to alkaline hydrogen peroxide or
- b)       reacting ammonia or a primary or secondary amine with an acid or an ester containing 1-3 structure elements according to formula II as defined in claims 1-6.

10           8.       Use of the compound according to claims 1-6 as a surfactant in a cleaning process.

9.       Use of the surfactant according to claim 8 for the cleaning of hard surfaces.

10.       Use of the surfactant according to claim 8 in a vehicle cleaning process, for bottle cleaning, machine dishwashing or machine washing of textiles.

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DECLARATION AND POWER OF ATTORNEY

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name.

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:  
**COMPOUNDS FROM EPOXIDISED, NITRILES, PROCESS FOR THEIR PRODUCTION AND USE AS CLEANING AGENTS**

was filed on **19 June 2000** as Appln. Ser. No. **PCT/SE00/01285**

I hereby state that I have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above. All factual statements made in the specification of my own knowledge are true and all factual statements made on information and belief are believed to be true.

I acknowledge to the duty to disclose information that is material to the patentability of this application in accordance with Title 37, Code of Federal Regulations, Sec. 1.56(a).

I hereby claim foreign priority benefits under Title 35, United States Code, Sec. 119 of any foreign application(s) for patent or inventor's certificate listed below and have also identified below any foreign application for patent or inventor's certificate having a filing date before that of the application on which priority is claimed:

Priority Claimed

<u>9902450-7</u>	<u>Sweden</u>	<u>29 JUNE 1999</u>	<u>X</u> Yes <u>     </u> No
(Number)	(Country)	(Day/Month/Year)	

I hereby claim the benefit under Title 35, United States Code § 119 of any provisional application(s) listed below:

Appln. Ser. No.	Country	Day/Month/Year
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I hereby claim the benefit under Title 35, United States Code, Sec. 120 of any United States application(s) listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States application in the manner provided by the first paragraph of Title 35, United States Code, Sec. 112, I acknowledge the duty to disclose material information as defined in Title 37, Code of Federal Regulations, Sec. 1.56(a) which occurred between the filing date of the prior application and the national or PCT international filing date of this application:

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(Appln. Ser. No.)	(Filing Date)	(Status: patented, pending, abandoned)
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POWER OF ATTORNEY: As a named inventor, I hereby appoint the following as my attorneys of record, with full power of substitution and revocation, to prosecute this application and to transact all business in the Patent Office:

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I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further, that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Sec. 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

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